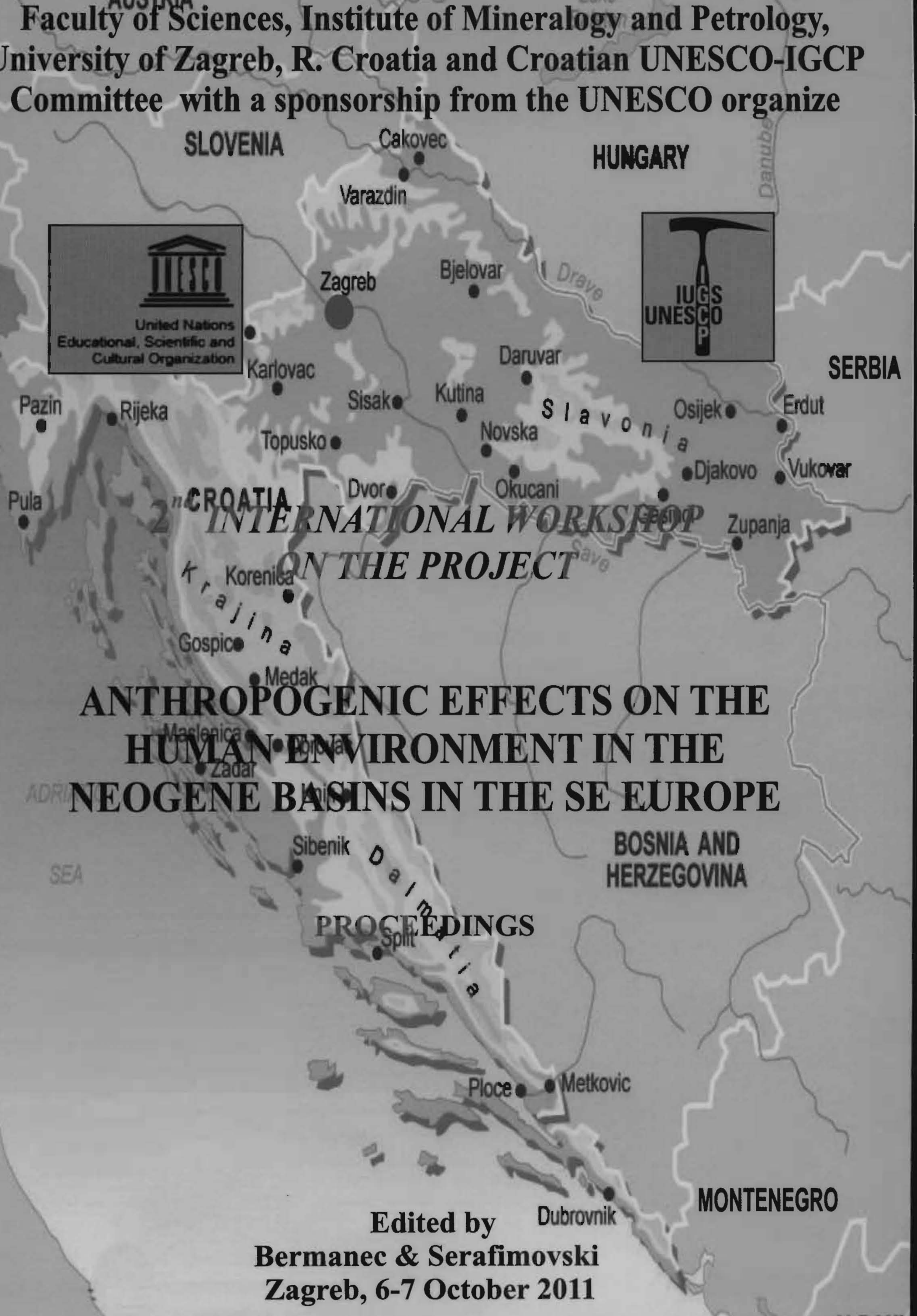
Faculty of Sciences, Institute of Mineralogy and Petrology, University of Zagreb, R. Croatia and Croatian UNESCO-IGCP Committee with a sponsorship from the UNESCO organize

2nd INTERNATIONAL WORKSHOP ON THE PROJECT

ANTHROPOGENIC EFFECTS ON THE HUMAN ENVIRONMENT IN THE NEOGENE BASINS IN THE SE EUROPE

PROCEEDINGS

Edited by
Bermanec & Serafimovski
Zagreb, 6-7 October 2011



RESULTS OF MEASUREMENTS OF SOIL ACTIVITY, SOIL GAS AND INDOOR RADON CONCENTRATIONS IN PRILEP AND SKOPJE

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Abstract: The preliminary measurements of radon and thoron concentrations in the soil gas were done, using short term active method. Field measurements were made at a distance of 1-2 m away from the randomly selected houses in Skopje and Prilep. Ten series of short-term (10-minutes) measurements were made. The arithmetic mean values of radon and thoron concentrations in the soil gas were found to be 15.9 ± 5.6 kBq m^{-3} and 5.3 ± 2.3 kBq m^{-3}, respectively. Activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil were evaluated from gamma spectrometry analysis on the soil samples that were collected from the same locations. Indoor radon concentrations were measured in the houses of the same locations.

1. Introduction

Radioactivity is a part of our everyday's life and as such, it is present in every medium of the living environment. The study of radioactivity in the living environment is a crucial segment of radiation protection, but at the same time, it is a useful tool in the exploration of the transport processes which originate from the nature itself.

In the period of 2007-2009, a survey of natural radioactivity was carried out in whole regions of Republic of Macedonia (1-6). Attempts were made in all investigates areas to identify populations receiving elevated natural exposures that might serve as potential groups for a planned future health study.

From human perspective, indoor radon (^{222}Rn) and its progeny are one of the most significant natural sources of radiation exposure to the population. As an inert gas, radon freely diffuses through the soils and reaches the atmosphere where it could migrate into structure of dwellings to pose a health hazard. The national survey of indoor radon concentration in 2008 was done. Indoor radon concentration was measured in 413 dwellings by using CR-39 detectors, with a total 1652 measurements. Detectors were deployed in four seasons in living room or bedroom of each house, where inhabitants spent the most of the time (5,7).

Some researches even believe that measurements of radon in the soil gas can be used to predict indoor radon level. In addition, numerous activities in the field of radon measurements in soil have been published (8-11). In order to give a more comprehensive evaluation of exposure and to identify sources of indoor radon concentrations in Skopje and Prilep, some measurements in 16 dwellings were undertaken in 2010. In this paper, we report the results of these measurements, with the emphasis on their correlations.

2. Study area

In order to investigate the concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil, radon and thoron concentrations in the soil gas and indoor radon concentrations, as well as to test its correlations, 16 testing sites were selected randomly for this study. These sites are situated a two towns Prilep and Skopje.

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3. Measuring procedure

3.1. Activity concentrations in the soil

The soil samples were collected on two different depths (0-20 cm and 20-100 cm). All samples were grinded and then dried at 105°C temperature until the moisture of the sample was completely evaporated. After homogenization, the samples were transferred in 500 ml Marinelli beaker which was used for the gamma spectrometry measurements. The containers were closed and sealed hermetically and kept aside for a month, in order to achieve a secular equilibrium between ^{226}Ra and its daughters.

The gamma spectrometry measurements were carried out with a p-type HPGe detector (Canberra Inc.; 25% relative efficiency, resolution of 1.79 keV at 1.33 MeV, 8192 ch. digital analyser, and with software GENIE 2000 for spectrum evaluation.

From the gamma spectrum the activity of the following radionuclides: ^{40}K, ^{226}Ra and ^{232}Th was determined. The activity of ^{40}K was determined from its 1460 keV line. The activity of ^{226}Ra was determined from the gamma lines associated with the short-lived daughters of: ^{214}Pb (295.22 keV, 351.93keV) and ^{214}Bi (609.31 keV, 1120.29 keV, 1764.49keV). Finally, the activity of ^{232}Th was determined from the gamma lines of ^{228}Ac (338.32 keV, 911.2 keV, 968.97 keV), ^{208}Tl (583.19 keV).

3.2. Indoor radon concentrations

The indoor radon concentration was measured at the same position locations during the four seasons (4 times a year with the same time of exposure, 3 months) in period 2008-2009.

The measurements were performed with commercially available RSKS passive track detectors. The detector consists of a CR-39 chip placed in a cylindrical diffusion chamber with dimensions 25 mm x 40 mm.

The detectors were set either in the living rooms or the bedrooms inside the houses, at a height of 1 to 1.5 m above the floor, at a distance greater than 0.5 m from each wall (to avoid influence of ^{220}Rn originating from the building materials, which contributes negligibly to the room interior, due to the low diffusion length) and, minimum 20 cm away from any other object in the room.

At the end of each seasonal cycle, the detectors were returned to the laboratory for analysis.

3.3. Radon and Thoron concentrations in soil gas

The measurement set-up consisted of the SARAD (RTM2100) radon monitor and soil gas sampling system. The RTM2100 - semiconductor detector combined with the alpha spectrometry analysis measured simultaneously radon and thoron concentrations by the short living daughter product (^{218}Po and ^{216}Po). The measurement locations were set at the house's courtyards for which the indoor concentrations of radon were measured as they were randomly selected, in Prilep and Skopje. Attention was payed that the selection was made from different town sections. The measurements in a selected location were made at a 1-2 m distance from each selected house.

A one meter (1 m) deep hole with 7 cm diameter was drilled. The soil sampler was carefully inserted into the bore-hole and connected directly to the instrument (connected to internal pump) by a flexible PVC tube. Ten series of short-term (10-minutes) measurements were made, and each result was recorded and kept for further processing.

4. Results and discussion

The results of the gamma spectrometry measurements of the soil samples, taken from holes on different depths (0-20 cm and 20-100 cm), are presented in Table 1. The activity concentration as well as the total combined uncertainty at the 68% confidence level of ^{40}K, ^{226}Ra and ^{232}Th are in Bq kg^{-1}.

The arithmetic mean (± standard deviation of the mean value) of the activity concentration of ^{40}K, ^{226}Ra and ^{232}Th on the (0 - 20)cm were (585 ± 33) Bq kg^{-1}, (38 ± 3) Bq kg^{-1} and (38 ± 2) Bq kg^{-1}, respectively. The activity of ^{226}Ra ranges from 19 to 56 Bq kg^{-1} and from 20 to 57Bq kg^{-1} on the (0-20)cm and (20 – 100)cm deeps respectively The range of measured activity on the (0-20)cm of ^{232}Th was 24-52 Bq kg^{-1} and 26-53 Bq kg^{-1} on the 20-100cm deep. The ranges of measured activity of ^{40}K on the (0-20)cm and (20-100)cm deeps were ranged from 319 to 761 Bq kg^{-1} and 332 to 807 Bq kg^{-1}, respectively.

Table 1. Activity concentraction of ^{40}K, ^{226}Ra and ^{232}Th in soil samples

N	Location	0-20cm						20-100cm					
		^{40}K		^{226}Ra		^{232}Th		^{40}K		^{226}Ra		^{232}Th	
		A*	u(A)**	A	u(A)	A	u(A)	A	u(A)	A	u(A)	A	u(A)
1	Prilep	761	10	53	0,4	49	0,6	765	10	54	0,7	53	0,6
2	Prilep	576	8.0	42	0,4	36	0,5	611	8,3	42	0,4	37	0,5
3	Prilep	749	9.9	42	0,4	41	0,5	807	10	41	0,4	39	0,5
4	Prilep	685	9,2	44	0,4	43	0,5	707	9,4	43	0,4	47	0,5
5	Prilep	659	9,2	47	0,4	42	0,5	728	9,6	57	0,5	51	0,6
6	Prilep	682	9,3	56	0,5	52	0,6	731	9,5	52	0,4	49	0,5
7	Prilep	609	11	47	0,6	47	0,8	555	7,8	49	0,4	45	0,5
8	Prilep	732	9,7	52	0,4	52	0,6	727	9,7	49	0,4	48	0,6
9	Prilep	559	7,7	44	0,4	47	0,5	569	7,7	46	0,4	53	0,5
10	Skopje	505	7,5	22	0,3	30	0,5	491	30	21	0,4	31	0,7
11	Skopje	604	8,5	30	0,3	30	0,4	715	9,7	25	0,3	34	0,5
12	Skopje	625	8,6	38	0,4	33	0,5	639	8,6	35	0,3	32	0,5
13	Skopje	434	6,2	21	0,2	29	0,4	457	6,6	25	0,2	32	0,4
14	Skopje	379	5,9	31	0,3	24	0,4	403	6,4	25	0,3	26	0,4
15	Skopje	319	6,1	19	0,2	26	0,4	332	5,4	20	0,2	28	0,4
16	Skopje	488	7,5	25	0,3	29	0,4	488	7,5	25	0,3	38	0,5

*activity concentration in Bq kg^{-1}.
**total combined uncertainty (σ=1) Bq kg^{-1}.

It seems useful to examine the downward distribution for ^{40}K, ^{226}Ra and ^{232}Th in surface soil to obtain an insight into mixing near the surface. For this purpose, radioactivity ^{40}K, ^{226}Ra and ^{232}Th, was compared for each sample taken from the two depths of each hole (12). A nonparametric linear regression analysis was performed, and the Spearman correlation coefficient was 0.94 for ^{40}K, 0.96 for ^{226}Ra and 0.87 for ^{232}Th, at the level of significance $p=0.05$.

From the statistical analysis, the results of the gamma spectrometry analysis of the soil samples, taken from holes, showed statistically insignificant differences between activity concentration of ^{40}K, ^{226}Ra and ^{232}Th from different depths (0-20cm and 20-100cm). On Figure 1 are presented the mean values of ^{226}Ra and ^{232}Th at 0 -100 cm depth.

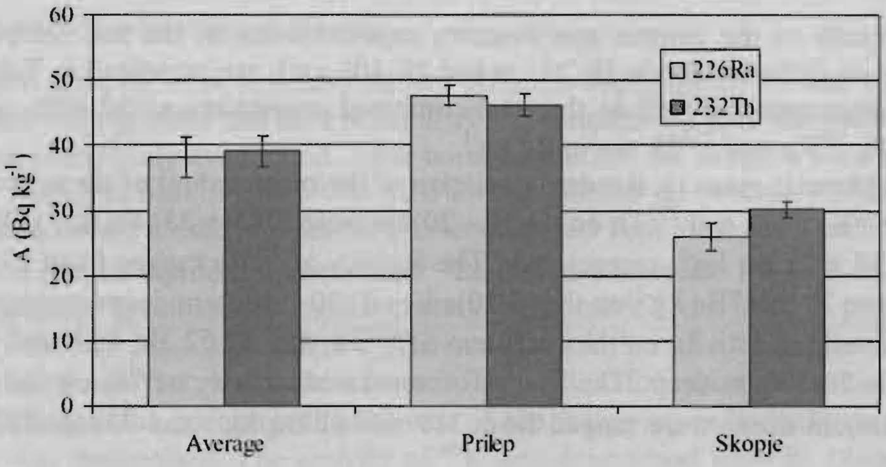

Figure 1. Mean values and 95% LSD intervals of the ^{226}Ra and ^{232}Th concentration at 0-100 cm depth

In Tabe 2, the results of indoor radon concentration measured in different season are present. The annual average indoor concentration is estimated as an arithmetic mean value of measured results in four seasons. All results are presented in Bq m^{-3}.

As can be seen from Table 2, the maximum value of radon concentration was 956 Bq m^{-3} and the minimum 16 Bq m^{-3} measured in winter and summer respectively. The indoor radon concentrations are subjected in seasonal variation. This could be attributed to the applied passive techniques, which are advantageous due to averaging of the seasonal variations caused by the long measurement period. The annual radon concentrations were ranged between 40 and 552 Bq m^{-3}.

Table 2. Results of indoor radon concentration ^{222}Rn

N	Location	^{222}Rn (Bq m^{-3})				
		Winter	Sping	Summer	Autumun	Annual average
1	Prilep	112	70	33	51	**66**
2	Prilep	300	221	117	198	**209**
3	Prilep	89	44	52	43	**57**
4	Prilep	117	95	41	69	**80**
5	Prilep	59	47	42	58	**51**
6	Prilep	323	178	73	154	**182**
7	Prilep	54	41	29	35	**40**
8	Prilep	72	79	64	43	**64**
9	Prilep	956	478	128	647	**552**
10	Skopje	45	51	16	50	**40**
11	Skopje	127	60	38	114	**85**
12	Skopje	335	157	171	301	**241**
13	Skopje	738	331	53	294	**354**
14	Skopje	291	173	38	293	**199**
15	Skopje	274	141	59	202	**169**
16	Skopje	104	49	39	94	**71**

The active method was used to determine the radon and thoron concentrations in the soil gas. The meteorological parameters: air temperature, pressure and relative humidity were measured with the same device because it is known that they can influence radon concentration. Therefore it seems important to know under what conditions the short-term radon measurements were carried out (13). The average values of air temperature, air pressure and relative air humidity ranged from 19 °C to 46 °C, 932 hPa to 980 hPa and 26% to 73%, respectively. The results of the soil gas measurements on the same locations are presented in Table 3.

Table 3. Radon and Thoron concentrations in soil gas

		^{222}Rn		^{220}Rn	
N	Location	C (kBq m^{-3})	u(C) (kBq m^{-3})	C (kBq m^{-3})	u(C) (kBq m^{-3})
1	Prilep	57.6	0.9	9.5	0.3
2	Prilep	23.4	0.4	5.2	0.2
3	Prilep	2.6	0.1	0.4	0.0
4	Prilep	3.3	0.1	2.5	0.1
5	Prilep	4.5	0.3	1.0	0.1
6	Prilep	3.7	0.2	0.5	0.1
7	Prilep	2.9	0.2	3.7	0.3
8	Prilep	2.8	0.1	2.2	0.1
9	Prilep	0.8	0.1	0.9	0.1
10	Skopje	20.7	0.4	38.6	0.1
11	Skopje	57.1	0.8	8.2	0.3
12	Skopje	62.5	0.7	7.3	0.2
13	Skopje	0.2	0.0	0.2	0.0
14	Skopje	6.1	0.2	1.4	0.1
15	Skopje	1.0	0.1	0.6	0.1
16	Skopje	5.0	0.2	2.5	0.1

The values of radon concentration in soil gas using short-term measurements obtainned using active devices varied from 0.2 kBq m^{-3} to 62.5 kBq m^{-3}. Obtained results for thoron concentrations in soil gas were found to vary from 0.2 kBq m^{-3} to 38.6 kBq m^{-3}. Nonparametric linear regression analysis of radon (^{222}Rn) and thoron (^{220}Rn) concentrations in soil gas has showed a statistically significant positive correlation with the Spearman correlation coefficient R=0.81 (p=0.0003). The mean values and 95% LSD intervals of the concentration of radon and thoron in soil gas are presented on the figure 2.

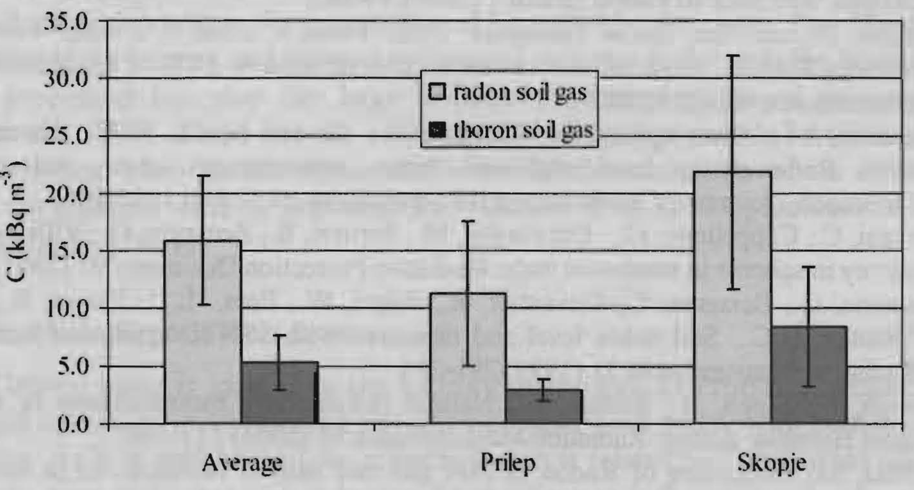

Fig. 2. Mean values and 95% LSD intervals of the radon and thoron concentration in soil gas

Furthermore the correlation between the ^{226}Ra content in the soil and the radon concentration in the soil gas were observed. There is no statistically significant correlation between radon concentration in soil gas and activity concentrations of ^{226}Ra in soil (p=0.805). Correlation between thoron (^{220}Rn) concentration in soil gas and soil activity concentrations of ^{232}Th, as well as correlation between the indoor radon concentration of and radon concentration in soil gas were statistically insignificant.

5. SUMMARY

A limited number of measurements of radon and thoron soil gas concentrations in the soil gas in Skopje and Prilep led to conclusion that there is a correlation between radon and thoron concentrations in soil gas, as well as a correlation between activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in soil is confirmed. Possibilities for other correlations are not excluded. However, the number of the data points is too small to allow generalization of the last conclusion. These results can be utilised to set up the methodology for a more systematic investigation to radon concentration in the soil gas.

6. REFERENCES

(1) Zdenka Stojanovska and Dusan Nedelkovski., Initial investigation of the natural radioactivity in the soil in some locations in Macedonia from radiation protection point of view. Physica Macedonica 58 (2008) 67-74.

(2) Димовска, С., 2007. Дистрибуција на некои природни и вештачки радионуклиди во почвата од Велес и неговата околина. Специјалистички труд, Хемиски институт, ПМФ, Скопје.

(3) Димовска, С., 2007. Радиоактивност на почвите од Кавадарци и неговата околина. Специјалистички труд, Хемиски институт, ПМФ, Скопје.

(4) Zdenka Stojanovska, Blazo Boev, Mimoza Ristova, Spatial Distribution of K, Th and U Elemental Concentrations in Soils of the Republic of Macedonia. 1-st International workshop on the UNESCO-IGCP Project (2011).

(5) Zdenka Stojanovska, Jovan Januseski, Blazo Boev, Mimoza Ristova,. Indoor exposure of population to radon in The FYR of Macedonia. Radiation Protection dosimetry (2011) 1-6.

(6) Zdenka Stojanovska, Nedelkovski Dusan, Jovan Janusevski.. Radon concentration in ground dwellings in winter/spring season and dose assessment in the citizens of Skopje. Archives of Public Health 1 (2009) 97-103.

(7) Zdenka Stojanovska, Jovan Januseski, Peter Bossew, Zora S. Zunic, Tore Tollefsen, Mimoza Ristova, Seasonal indoor radon concentration in FYR of Macedonia. Radiation measurement 46, (2011)602-610.

(8) Ngachin, M., Garavaglia, M., Giovani, C., Kwato Njock, M.G., Nourreddine, A., 2008. Radioactivity level and soil radon measurement of a volcanic area in Cameroon. Journal of Environmental Radioactivity 99 (2008) 1056-1060.

(9) Giovani, C., Cappelletto, C., Garavaglia, M., Scruzzi, E., Peressini, G., Villalta, R.,. Radon survey in schools in north-east Italy. Radiation Protection Dosimetry 97 (2001) 341-344.

(10) Jonsson, G., Baixeras, C., Devantier, R., Engel, W., Font,. L.L., Freyer, K., Ghose, R., Theutler, H.C.,. Soil radon level and measured with SSNTD's and soil radium content. Radiation Measurements 31 (1999) 291-294.

(11) Singh, S., Singh, B., Kumar, A., Natural radioactivity measurements in soil samples from Hamipur district. Radiation Measurements 36 (2003) 547-549.

(12) Fukui, M. Evaluation of Radon in Soil gas and natural radioactivity in surface soil of Kinki District, Japan. Journal of nuclear science and technology, 44 (2007) 1106-1116.

(13) Zunic, Z. S., Kobal, I., Vaupotič, J., Kozak, K., Mazur, J., Birovljev, A., Janik, M., Celikovicr , I., Ujic, Demajo, A., Krsticr , G., Jakupi, B., Quarto, M., Bochicchio, F.,. High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan Region). J. Environ. Radioact. (2006) 89, 249–26.